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RADIOCARBON CALIBRATION FOR JAPANESE WOOD SAMPLES

Minoru Sakamoto¹ • Mineo Imamura^{1,2} • Johannes van der Plicht³ • Takumi Mitsutani⁴ • Makoto Sahara^{1,5}

ABSTRACT. The radiocarbon content of Japanese cedars was measured by accelerator mass spectrometry for decadal tree-ring samples from the period of 240 BC to AD 900. Conventional gas counting was also used for part of the samples. The data were compared with the INTCAL98 calibration curve (Stuiver et al. 1998). The results indicate that the difference in atmospheric ¹⁴C between Japan and North America or Europe is negligible at this period, less than 18 ¹⁴C yr using an average of 50 yr. However, in the period of about AD 100 to about AD 200, we cannot exclude the possibility of a deviation of the order of 30 to 40 ¹⁴C yr to the older ages.

INTRODUCTION

Precise calibration data are available for the atmospheric ¹⁴C record of the past, enabling us to estimate true ages with much more confidence than before. The combination of ¹⁴C dates and dendrochronology can lead to an exact age determination. Moreover, accurate dates—up to about 10–20 calibrated yr—can be obtained by means of the so-called wiggle-matching technique. Therefore, precise dating by ¹⁴C is of increasing interest for archaeologists. Precise knowledge on the local effect of the atmospheric ¹⁴C content is becoming more important for future perspectives of precise ¹⁴C dating.

Thus far, conversion of ¹⁴C dates to calendar yr has been based on ¹⁴C calibration curves obtained from North American and European trees for Japanese samples. No calibration curve for Asian trees is available. It is important to examine possible regional deviation from INTCAL98 and investigate the extent of the regional effect in ¹⁴C calibration. By doing this, we can safely apply the INTCAL98 calibration curve to the Japanese samples. The aim of this research is to obtain precise ¹⁴C dates for dendrochronologically-dated woods, particularly for the Yayoi and Kofun periods, which are described below.

For archaeological research in Japan, the period of several centuries around AD 1 has been of great interest because in this period rapid changes took place in many cultural and social aspects. The first half of the period corresponds to the prehistoric era (Yayoi period, 4th century BC to 3rd century AD) during which paddy rice farming has spread over the islands and the use of bronze and iron started. The latter half corresponds to the protohistoric era (Kofun period, 3rd century AD to 6th century AD), which is characterized by huge burial mounds and the establishment of the first state in Japan. It is also conceived that cultural exchange was quite active between the East Asian continent and the Japanese archipelago. The absolute chronology of these periods is controversial, since recent dendrochronological dates obtained for several Yayoi archaeological sites show a significant discrepancy from the archaeological chronology that had been widely accepted (Mitsutani 2002). Therefore, precise dating has been of great concern for many archaeologists in Japan as well. Recent studies show that ¹⁴C dating within a few tens of yr is possible using the wiggle-matching technique, as mentioned above. The use of the universal calibration curves for Japanese wood samples should be suitable for practical use, however, it is necessary to prove that the curves are correct.

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We have measured the ^{14}C dates of decadal tree rings using 4 Japanese wood specimens that have been dendro-dated. By comparing ^{14}C dates with the INTCAL98 calibration curve, we discuss in detail the applicability of INTCAL98 to the precise ^{14}C dating of the pre- and proto-historic periods in Japan. This produces the first contribution to a calibration curve for Japan. Because of the limited amount of wood available, most of the measurements were done by AMS. Eleven measurements were done by the conventional method.

DESCRIPTIVE BACKGROUND

Sample Source and Tree-ring Dating

Four Japanese cedar (*Cyptomeria japonica*) specimens—HK, MT, AH1, and AH3—were selected for this study. HK was a bogwood unearthed at Hakone, Kanagawa Prefecture, central Honshu Island located about 100 km west of Tokyo. The ring patterns of HK spanned 453 yr. MT was also a bogwood unearthed at Miyata-mura village, Nagano Prefecture, central Honshu Island, with total ring patterns of 372 yr. AH1 and AH3 were individual piles excavated at the Hotta-no-saku archaeological site in Akita Prefecture, northern Honshu Island. The locations of these samples are shown in Figure 1.

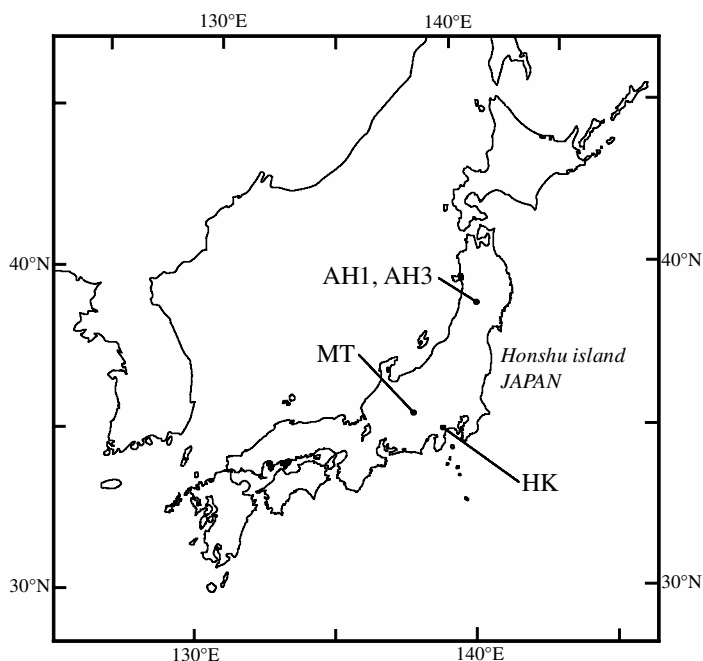


Figure 1 Map of sample locations

Dendrochronological determination of the absolute age for these specimens was performed by comparing the standardized ring patterns with the master chronology for Japanese cedar developed by the Nara National Culture Properties Research Institute (1990). A master standard chronology for a Japanese cedar has been established for tree-ring ages back to 1313 BC.

Each specimen was cut into decadal pieces according to its dendro-date. Although the ring width varied, we assumed that each decadal piece represents the mean ^{14}C concentration in that time

period. Thirty-two samples were obtained from HK corresponding to the age of 240 BC to AD 200, 30 samples from MT (AD 331 to AD 630), 11 samples from AH1 (AD 692 to AD 801), and 24 samples from AH3 (AD 661 to AD 900), respectively. Preliminary measurement using the HK specimen for 201 BC to AD 60 was reported by Imamura et al. (1998).

The surface portion of the sample was removed to prevent possible contamination with modern carbon, and each decadal sample piece was pulverized into fine fiber tips of about 0.1×1 mm size using a mill (an electric blender).

¹⁴C Measurements

Accelerator Mass Spectrometry (AMS) (lab. code GrA)

The HK, MT, and AH3 samples were measured for ¹⁴C using AMS. About 100 mg of the sample fiber was treated by acid-alkali-acid (in each step, the sample was reacted 5 times with 1.2N HCl or 1.2N NaOH for 1 hr), followed by removal of lignin with Cl₂ (using NaClO₂ and HCl) and most β- and γ- cellulose with a 17.5% NaOH solution. The purified sample, mainly consisting of α-cellulose, was then neutralized, washed with pure H₂O, filtrated, and dried.

Several mg of cellulose were taken in a Vycor-glass tube together with few hundred mg of CuO (organic carbon analysis grade: Wako Chemical Co.) and sealed off from the vacuum system. The sample tube was then heated at 850 °C for 2 hr to completely oxidize the cellulose. The obtained CO₂ was transferred to the high-vacuum CO₂ purification systems (at the National Museum of Japanese History for HK and AH3, and at the University of Groningen for MT) and purified cryogenically. The pressure of the purified CO₂ was measured by a gauge and divided into 2 breakseals. Graphite targets were prepared in Groningen and the ¹⁴C/¹²C and ¹³C/¹²C ratios were measured with the Groningen HVEE AMS system (van der Plicht et al. 2000). ¹⁴C dates were determined from the ¹⁴C/¹²C ratios after normalizing the isotopic ratio of ¹³C/¹²C to $\delta^{13}\text{C} = -25.0$ per mil.

Conventional ¹⁴C measurements (lab. code GrN)

The AH1 samples were measured for ¹⁴C in the Groningen conventional laboratory. Several g of the sample fiber were treated by the acid-alkali-acid method. Treated fiber was combusted into CO₂ and purified. Obtained CO₂ was collected in a metal cylinder, introduced into a proportional counter, and measured for ¹⁴C β-rays radiation (Mook and Streurman 1983; van der Plicht et al. 1992). Also, here the ¹⁴C dates are reported including a correction for fractionation to $\delta^{13}\text{C} = -25.0$ per mil.

DISCUSSION

The results of the ¹⁴C measurements for each sample specimen are shown in Tables 2–5. The data include our preliminary report on the 200–60 BC samples for HK (Imamura et al. 1998). The errors include the statistics of the ¹⁴C counts and uncertainties in the ¹³C/¹²C and ¹⁴C/¹²C ratios of the standard and blank targets. The extent of modern carbon contamination was of the order of 0.1% or less, which is negligible. Uncertainties of original ¹⁴C standards are not included. In Figure 2, the results are compared with the calibration curve of INTCAL98 (Stuiver et al. 1998). Error bars represent a 68% (1σ) uncertainty.

From the data plotted in Figure 2, it is shown that almost all the data are in good agreement (within 2σ) with the INTCAL98 calibration curve. There may be an exception in the period of around AD 100–200, which may show a significant deviation toward the older ages and will be discussed later in more detail.

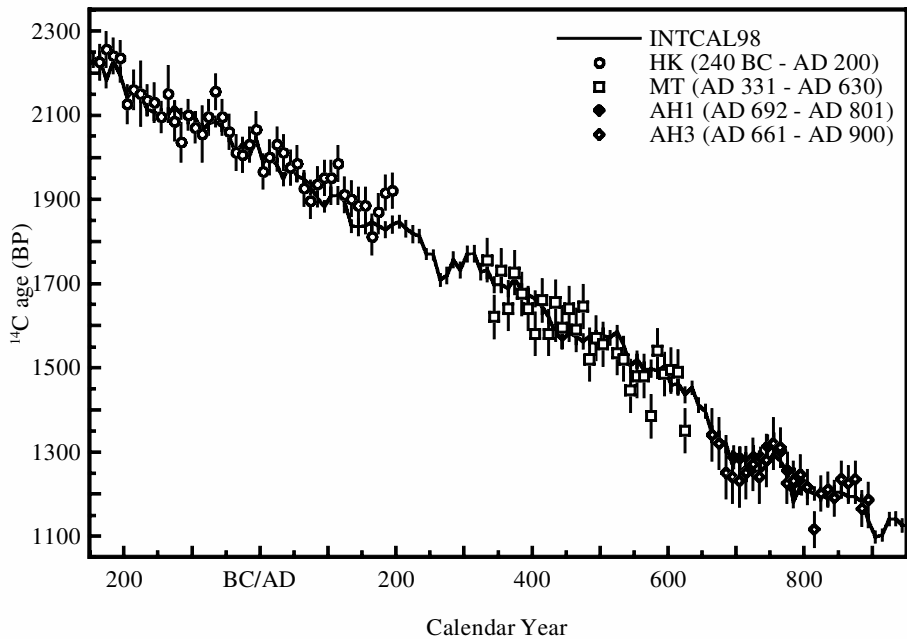


Figure 2 ^{14}C data for Japanese cedars compared with INTCAL98

Table 1 Source of the tree-ring sample specimens

Sample code	Locality	Latitude	Longitude	Tree-ring age
HK	Hakone-machi Town, central Honshu Island, Japan	35°10'N	139°05'E	240 BC–AD 200
MT	Miyata-mura Village, central Honshu Island, Japan	35°45'N	137°55'E	AD 331–AD 630
AH1	Hotta-no-saku archaeological site, northern Honshu Island, Japan	39°30'N	140°35'E	AD 692–AD 801
AH3	Hotta-no-saku archaeological site, northern Honshu Island, Japan	39°30'N	140°35'E	AD 661–AD 900

The significance of deviation for each datum from the INTCAL98 can be given by the formula:

$$\Delta Z = \frac{T(t)_s - T(t)_{\text{INTCAL98}}}{\sqrt{\sigma_s^2 + \sigma_{\text{INTCAL98}}^2}}$$

where ΔZ is the normalized difference, $T(t)_s$ and $T(t)_{\text{INTCAL98}}$ are the ^{14}C ages of the decadal tree-ring sample and the corresponding INTCAL98 curve for a calibrated age, t , and σ_s , σ_{INTCAL98} are their respective uncertainties. The ΔZ values are calculated for the individual dataset of each specimen. The distribution of cases is plotted in Figure 3 as a function of ΔZ . The total number of the case is normalized to 1. Figure 3 shows the distribution for each sample specimen is nearly a typical normal distribution and the average of the distribution is very close to zero. This is what is expected when there are no biases in the measurements and no significant regional effect for the whole period investigated. By calculating a best fit to a normal distribution, the shifts are calculated to be 0.42, -0.23,

Table 2 Results of ^{14}C measurements (AMS) for the HK series

Lab code	Sample name	Tree-ring age	^{14}C age (BP)
GrA-14700	HK-235B	240 BC – 231 BC	2225 \pm 40
GrA-14702	HK-225B	230 BC – 221 BC	2255 \pm 40
GrA-14703	HK-215B	220 BC – 211 BC	2240 \pm 40
GrA-14704	HK-205B	210 BC – 201 BC	2235 \pm 40
GrA-9332	HK-195B	200 BC – 191 BC	2125 \pm 45
GrA-9326	HK-185B	190 BC – 181 BC	2160 \pm 45
GrA-8085	HK-175B	180 BC – 171 BC	2150 \pm 75
GrA-9323	HK-165B	170 BC – 161 BC	2090 \pm 45
GrA-9330	HK-165B	170 BC – 161 BC	2180 \pm 45
GrA-9328	HK-155B	160 BC – 151 BC	2130 \pm 45
GrA-8082	HK-145B	150 BC – 141 BC	2130 \pm 65
GrA-9419	HK-145B	150 BC – 141 BC	2080 \pm 40
GrA-8081	HK-135B	140 BC – 131 BC	2150 \pm 65
GrA-9322	HK-125B	130 BC – 121 BC	2085 \pm 45
GrA-9327	HK-115B	120 BC – 111 BC	2035 \pm 45
GrA-9316	HK-105B	110 BC – 101 BC	2080 \pm 45
GrA-9317	HK-105B	110 BC – 101 BC	2120 \pm 45
GrA-9318	HK-095B	100 BC – 91 BC	2085 \pm 45
GrA-9321	HK-095B	100 BC – 91 BC	2050 \pm 45
GrA-8084	HK-085B	90 BC – 81 BC	2055 \pm 65
GrA-14705	HK-075B	80 BC – 71 BC	2095 \pm 40
GrA-14707	HK-065B	70 BC – 61 BC	2155 \pm 40
GrA-14709	HK-055B	60 BC – 51 BC	2095 \pm 40
GrA-14689	HK-045B	50 BC – 41 BC	2060 \pm 40
GrA-14688	HK-035B	40 BC – 31 BC	2010 \pm 40
GrA-14627	HK-025B	30 BC – 21 BC	2005 \pm 40
GrA-14628	HK-015B	20 BC – 11 BC	2030 \pm 40
GrA-14629	HK-005B	10 BC – 1 BC	2065 \pm 40
GrA-14630	HK-005A	AD 1 – AD 10	1965 \pm 40
GrA-14632	HK-015A	AD 11 – AD 20	2000 \pm 40
GrA-14684	HK-025A	AD 21 – AD 30	2030 \pm 40
GrA-14685	HK-035A	AD 31 – AD 40	2010 \pm 40
GrA-14690	HK-045A	AD 41 – AD 50	1975 \pm 40
GrA-14691	HK-055A	AD 51 – AD 60	1985 \pm 40
GrA-14693	HK-065A	AD 61 – AD 70	1925 \pm 40
GrA-14694	HK-075A	AD 71 – AD 80	1895 \pm 40
GrA-14695	HK-085A	AD 81 – AD 90	1935 \pm 40
GrA-14698	HK-095A	AD 91 – AD 100	1950 \pm 40
GrA-14699	HK-105A	AD 101 – AD 110	1950 \pm 40
GrA-14710	HK-115A	AD 111 – AD 120	1985 \pm 40
GrA-14712	HK-125A	AD 121 – AD 130	1910 \pm 40
GrA-14713	HK-135A	AD 131 – AD 140	1900 \pm 40
GrA-14714	HK-145A	AD 141 – AD 150	1885 \pm 40
GrA-14715	HK-155A	AD 151 – AD 160	1885 \pm 40
GrA-14717	HK-165A	AD 161 – AD 170	1810 \pm 40
GrA-14718	HK-175A	AD 171 – AD 180	1870 \pm 40
GrA-14720	HK-185A	AD 181 – AD 190	1915 \pm 40
GrA-14722	HK-195A	AD 191 – AD 200	1920 \pm 40

Table 3 Results of ^{14}C measurements (AMS) for the MT series

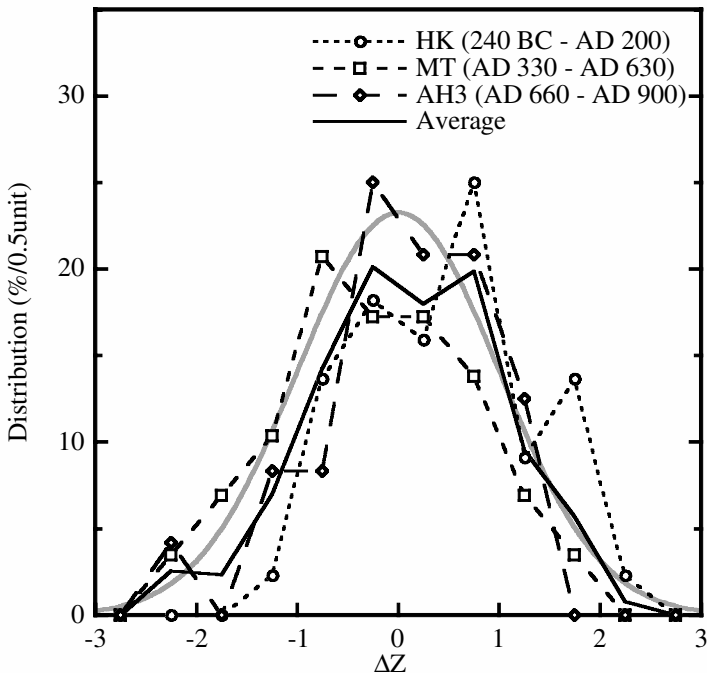
Lab code	Sample name	Tree-ring age	^{14}C age (BP)
GrA-15263	MT-335	AD 331 – AD 340	1755 ± 50
GrA-15264	MT-345	AD 341 – AD 350	1620 ± 50
GrA-15265	MT-355	AD 351 – AD 360	1730 ± 50
GrA-15266	MT-365	AD 361 – AD 370	1640 ± 50
GrA-15268	MT-375	AD 371 – AD 380	1725 ± 50
GrA-15269	MT-385	AD 381 – AD 390	1675 ± 50
GrA-15270	MT-395	AD 391 – AD 400	1640 ± 50
GrA-15273	MT-405	AD 401 – AD 410	1580 ± 50
GrA-15320	MT-415	AD 411 – AD 420	1660 ± 50
GrA-15275	MT-425	AD 421 – AD 430	1580 ± 50
GrA-15276	MT-435	AD 431 – AD 440	1655 ± 50
GrA-15278	MT-445	AD 441 – AD 450	1595 ± 50
GrA-15279	MT-455	AD 451 – AD 460	1640 ± 50
GrA-15280	MT-465	AD 461 – AD 470	1590 ± 50
GrA-15283	MT-475	AD 471 – AD 480	1645 ± 50
GrA-15287	MT-485	AD 481 – AD 490	1520 ± 50
GrA-15289	MT-495	AD 491 – AD 500	1570 ± 50
GrA-15291	MT-505	AD 501 – AD 510	1555 ± 50
GrA-24950	MT-515	AD 511 – AD 520	1480 ± 50
GrA-15293	MT-525	AD 521 – AD 530	1535 ± 50
GrA-15294	MT-535	AD 531 – AD 540	1520 ± 50
GrA-15296	MT-545	AD 541 – AD 550	1445 ± 50
GrA-15298	MT-555	AD 551 – AD 560	1480 ± 50
GrA-15299	MT-565	AD 561 – AD 570	1480 ± 50
GrA-15301	MT-575	AD 571 – AD 580	1385 ± 50
GrA-15302	MT-585	AD 581 – AD 590	1540 ± 50
GrA-15303	MT-595	AD 591 – AD 600	1485 ± 50
GrA-15304	MT-605	AD 601 – AD 610	1495 ± 50
GrA-15306	MT-615	AD 611 – AD 620	1490 ± 50
GrA-15307	MT-625	AD 621 – AD 630	1350 ± 50

Table 4 Results of ^{14}C measurements (conventional) for the AH1 series

Lab code	Sample name	Tree-ring age	$\delta^{13}\text{C}$ (‰)	^{14}C age (BP)
GrN-25080	AH1-696	AD 692 – AD 701	-26.35	1287 ± 25
GrN-25081	AH1-706	AD 702 – AD 711	-26.18	1284 ± 25
GrN-25082	AH1-716	AD 712 – AD 721	-26.42	1255 ± 25
GrN-25083	AH1-726	AD 722 – AD 731	-26.85	1287 ± 26
GrN-25084	AH1-736	AD 732 – AD 741	-26.57	1286 ± 23
GrN-25085	AH1-746	AD 742 – AD 751	-26.37	1313 ± 26
GrN-25086	AH1-756	AD 752 – AD 761	-26.56	1317 ± 26
GrN-25087	AH1-766	AD 762 – AD 771	-26.34	1301 ± 26
GrN-25088	AH1-776	AD 772 – AD 781	-25.58	1255 ± 43
GrN-25089	AH1-786	AD 782 – AD 791	-25.70	1228 ± 31
GrN-25090	AH1-796	AD 792 – AD 801	-25.37	1227 ± 28

Table 5 Results of ^{14}C measurements (AMS) for the AH3 series

Lab code	Sample name	Tree-ring age	^{14}C age (BP)
GrA-14997	AH3-665	AD 661 – AD 670	1340 ± 60
GrA-14998	AH3-675	AD 671 – AD 680	1320 ± 60
GrA-14999	AH3-685	AD 681 – AD 690	1250 ± 60
GrA-15000	AH3-695	AD 691 – AD 700	1240 ± 60
GrA-15002	AH3-705	AD 701 – AD 710	1230 ± 60
GrA-15003	AH3-715	AD 711 – AD 720	1250 ± 60
GrA-15004	AH3-725	AD 721 – AD 730	1270 ± 60
GrA-15007	AH3-735	AD 731 – AD 740	1240 ± 60
GrA-15008	AH3-745	AD 741 – AD 750	1280 ± 60
GrA-15009	AH3-755	AD 751 – AD 760	1320 ± 60
GrA-15082	AH3-765	AD 761 – AD 770	1310 ± 45
GrA-15084	AH3-775	AD 771 – AD 780	1225 ± 45
GrA-15086	AH3-785	AD 781 – AD 790	1230 ± 45
GrA-15095	AH3-795	AD 791 – AD 800	1245 ± 45
GrA-15139	AH3-805	AD 801 – AD 810	1215 ± 40
GrA-15140	AH3-815	AD 811 – AD 820	1115 ± 40
GrA-15143	AH3-825	AD 821 – AD 830	1200 ± 40
GrA-15144	AH3-835	AD 831 – AD 840	1210 ± 40
GrA-15146	AH3-845	AD 841 – AD 850	1190 ± 40
GrA-15153	AH3-855	AD 851 – AD 860	1235 ± 40
GrA-15150	AH3-865	AD 861 – AD 870	1225 ± 40
GrA-15151	AH3-875	AD 871 – AD 880	1235 ± 40
GrA-15152	AH3-885	AD 881 – AD 890	1165 ± 40
GrA-15155	AH3-895	AD 891 – AD 900	1185 ± 40

Figure 3 Difference between ^{14}C data for Japanese cedars and INTCAL98

and 0.16 for HK, MT and AH3, respectively. The data of AH1 are too small to show a meaningful distribution. If we take a typical error of each determination, the shifts are 18, -10, and 6 ^{14}C yr, respectively.

The wiggle-matched date of each specimen is calculated using all the data by comparing with the INTCAL98 calibration curve. The calculated dates are in very good agreement with the dendrochronological dates determined for each specimen. Although it is known that the calculated age should be less influenced by the regional effect (Bronk Ramsey et al. 2001), the observations above also enforce the correctness of using INTCAL98 calibration curve for Japanese wood samples.

The above conclusion is based on the data set of each sample specimen of a rather long period. To search the detailed structure of the regional effect, 50 yr of averages of the differences between the data and INTCAL98 are calculated and plotted in Figure 4. The figure shows the difference is within the 2σ uncertainties except for the values around AD 100 to AD 200, indicating absence of regional effect that exceeds 20 ^{14}C yr in the timescale of 50 calendar yr.

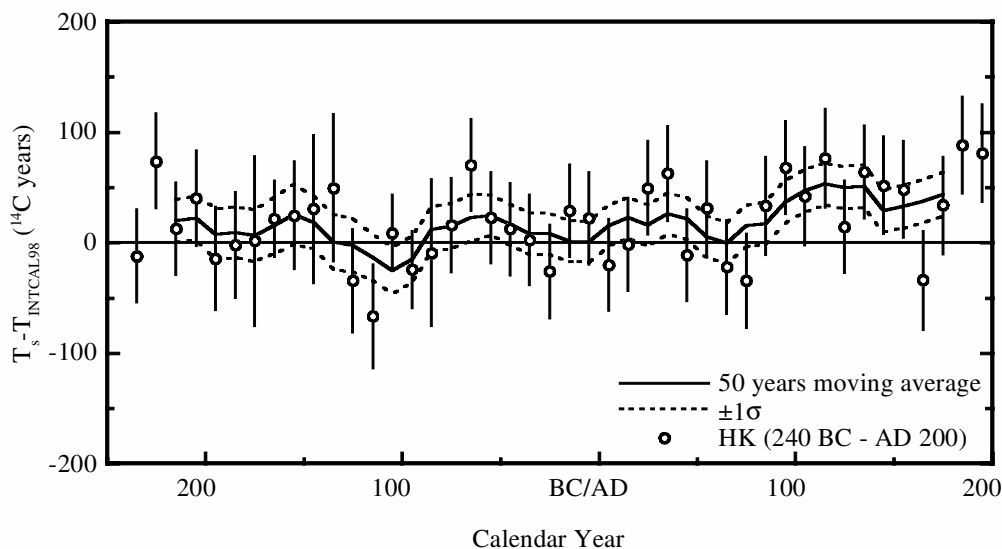


Figure 4 Difference of ^{14}C ages between Japanese cedar and INTCAL98

For the data of AD 100 to AD 200, we possibly observe a regional or local effect. The 50-yr averages for these periods certainly deviate more than the 2σ uncertainties from those of the INTCAL98. At present, we consider 2 possibilities: that the regional effect exists in this period for Japanese islands, or that the effect is only local due to the volcanic activities from a nearby volcano (Mt. Fuji for example) and restricted to the area around the Fuji-Hakone National Park in Japan where the HK sample was taken. We plan to measure another tree-ring sample of the same period but taken from a different area of Japan.

CONCLUSIONS

From the present study, it is clear that the differences in atmospheric ^{14}C concentrations between Japan and North America or Europe are negligible or small during the period of 270 BC to AD 900, suggesting that there is little disturbance of ^{14}C in the atmosphere caused by the ocean near the Japanese archipelago. This is likely because the air mass moves eastwards at the mid-latitude and

reaches Japan with little influence of the ocean. However, we found a possible deviation in the period of about AD 100 to AD 200, which requires future investigation.

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